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The absolute coherence times depend on several experimental parameters such as crystal purity, sample temperature, laser power, etc. As a result of changes in these parameters we find variations of the hole spin coherence time from about 200 ns up to 1 μ s, measured by the spin mode-locking effect in the quantum dot ensemble (without implementing spin echo protocols).

With increasing complexity of the laser-pulse protocol, the number of possible variation parameters in excitation increases. In the best possible implementation of the used setup that allows also application of multiple periodic pulses for dynamic decoupling we achieved

coherence times of about 600 ns. If these optimized conditions are not met, the coherence times can be considerably shorter as in the first version of the manuscript where mode-locking quickly disappeared with increasing pump separation.

Irrespective of the concrete conditions, the dynamic decoupling protocol with a separation of 13.2 ns between the pulses for spin inversion rendered coherence times comparable to or slightly longer than the coherence time without such protocols. Furthermore, the coherence times increased by a factor of about 2 when the protocol with 6.6 ns inversion pulse separation was applied.

All-optical implementation of a dynamic-decoupling protocol for hole spins in (In,Ga)As quantum dots

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We demonstrate the potential of a periodic laser-pulse protocol for dynamic decoupling of hole spins in (In,Ga)As quantum dots from surrounding baths. When doubling the repetition rate of inversion laser pulses between two reference pulses for orienting the spins, we find that the spin coherence time is increased by a factor of two.

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Solid state implementations of quantum information technologies have been considered attractive because of their potential advantages such as robustness, miniaturization, scalability and connection to conventional information processing hardware.^{1,2} The huge obstacle in such approaches is the 'rigid' coupling of the quantum bits to their surrounding, strongly limiting their coherence. The demand for long-lived coherence has quickly geared activities towards carrier spins in crystals.^{3,4} For them, carrier localization suppresses decoherence mechanisms involving orbital motion.⁵ This has led to the suggestion of the hyperfine interaction with the surrounding nuclear spin bath as the main decoherence mechanism for carrier spins.⁶⁻⁸

For further improvement in this respect, two different strategies can be pursued: either purification towards zero nuclear spin isotopes or implementation of protocols for dynamic decoupling from baths. Also a combination of both strategies may be applied, which has lead, for example, to an extension of the spin-coherence time associated with the NV⁻ center in diamond to milliseconds, even at room temperature.^{9,10} For III-V semiconductors isotope purification is not possible, but dynamic decoupling has shown great promise. For gate-defined quantum dots the coherence time could be extended at cryogenic temperatures from a few up to 200 μ s in that way.¹¹

Recently, considerable efforts have been made to develop optimized pulse sequences reaching a complexity far beyond the initially applied periodic Carr-Purcell-Meiboom-Gill (CPMG) protocol.¹²⁻¹⁷ This possibility is offered by the accurate electronic control of radiation pulses in the microwave frequency range. However, these pulses are limited to durations of more than a nanosecond. Much shorter pulses are possible employing lasers, but the possibility to vary the pulse properties within a manipulation sequence is limited. Due to these difficulties an extension of the spin-coherence time by dynamical decoupling with laser pulses has not yet been accomplished to the best of our knowledge, and this is the goal that we target here. Dynamic decoupling leads to an insensitivity to noise sources characterized by coupling frequencies lower than the pulse rate in the implemented protocol, opening up a frequency gap in the interaction

with surrounding baths.^{18,19} Therefore, implementations using pulses as short as possible are appealing because of the higher possible pulse rates that may be applied. Under these circumstances the spins become less sensitive to noise sources with enhanced coupling frequencies.

For that purpose we monitor the precession of hole spins confined in an (In,Ga)As/GaAs quantum dot ensemble about an external magnetic field after their orientation normal to that field by pump pulses.²⁰ To study the spin coherence, we optically stimulate spin echoes by applying rotation pulses, which invert the inhomogeneous dephasing of the hole spin ensemble precession. The underlying basic operation of a single π -rotation of spins was demonstrated earlier.^{21,22} From the echo amplitude dependence on the time after spin initialization the hole spin coherence time can be accurately assessed. The echo-inducing pulses are applied periodically, mimicking the CPMG protocol used for extending spin coherence in dynamical decoupling.²³ By doubling the repetition rate of rotation pulses we find that the hole spin coherence time is increased by a factor of about 2, underlining the possibility to implement dynamical decoupling also purely optically. In addition, we find that the spin-coherence dynamics shows signatures of a deviation from a simple exponential decay.

The experiments are performed on an ensemble of self-assembled (In,Ga)As/GaAs QDs, which show a resident hole occupation due to residual doping by carbon impurities, as demonstrated earlier.²⁰ The sample contains ten layers of QDs separated by 100-nm GaAs barriers, each with a dot density of 10^{10} cm^{-2} . The ground state emission maximum of the photoluminescence (PL) is at 1.38 eV with a full width at half maximum of 20 meV. The sample is mounted in a cryostat, where it is cooled to temperature $T = 6 \text{ K}$ and exposed to a magnetic field of $B = 1 \text{ T}$ along the x direction, normal to the sample growth direction that coincides with the optical axis z (Voigt geometry).

The hole spin dynamics is studied by a degenerate pump-probe setup employing in the simplest version of the experiment a pulsed Ti:Sapphire laser with its photon energy tuned to the PL maximum. The laser emits pulses with a duration of 1 ps at a repetition rate of 75.75 MHz.

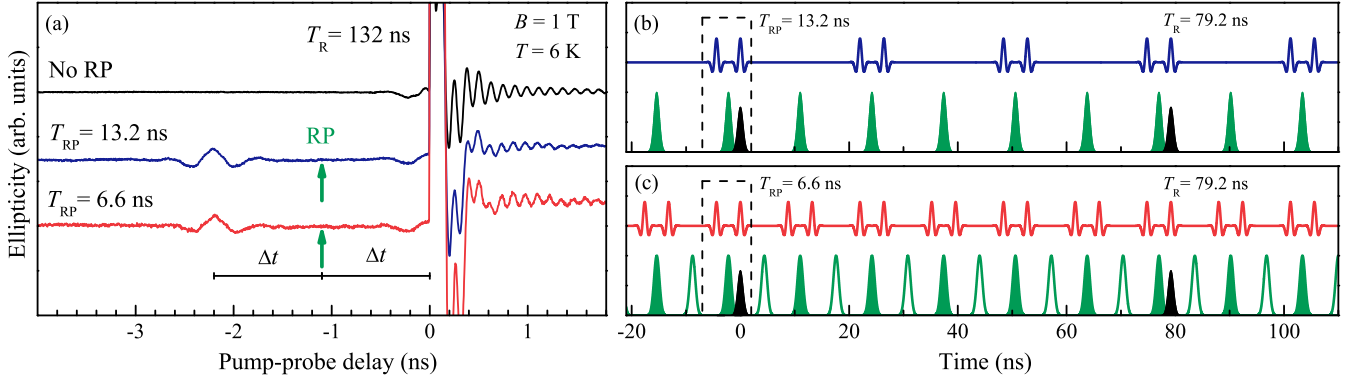


FIG. 1. (Color online) (a) Time-resolved ellipticity measurements at $B = 1$ T with pump-probe repetition period $T_R = 132$ ns at $T = 6$ K. The black, top trace shows the spin polarization around the pump incidence at zero delay without applying RPs. The middle, blue trace shows a measurement with an additional RP train shifted by 1.1 ns to earlier times (see the green arrow) relative to the pump train with a RP period $T_{RP} = 13.2$ ns, resulting in a hole spin echo 2.2 ns before pump incidence. In the lower, red trace the RP period is reduced to $T_{RP} = 6.6$ ns so that the number of rotations between two pump pulses is doubled compared to the 13.2 ns RP period. (b) Scheme of RP application (green) and echo appearance (blue) for $T_{RP} = 13.2$ ns between two pump pulses (black) with $T_R = 79.2$ ns. (c) Echo appearance (red) for $T_{RP} = 6.6$ ns. The additional RP incidences are indicated by the open green pulses.

The pulse period used for the experiment was reduced by a pulse picker letting every n -th pulse pass while blocking all other pulses in between such that the pulse repetition period T_R is a multiple of the original period of 13.2 ns. In our studies it was varied from 132 to 462 ns. The laser output is split into circularly polarized pump and linearly polarized probe pulses, which can be delayed relative to each other. The pump power is adjusted to a pulse area of $\Theta = \pi$,²² the probe power is about ten times weaker.

The train of pump pulses creates a spin polarization along the optical axis by exciting positively charged excitons (trions). After their radiative decay, spin-oriented holes are left behind, which subsequently precess about the magnetic field in the yz -plane. Before the decay, also precession of the optically excited electron spins contributes to the coherent signal. The spin polarization is monitored by measuring the ellipticity of the probe pulses acquired by transmission through the sample. The upper black trace in Fig. 1(a) shows a time-resolved ellipticity measurement of the spin polarization with pump incidence at zero pump-probe delay. After the incidence one observes dominantly fast precession from photoexcited electrons and superimposed slow spin precession from the resident holes, each corresponding to the characteristic g factors. The observed number of hole spin oscillations is rather small due to fast dephasing, arising from the considerable g factor inhomogeneity in the ensemble. The decay of the electron spin precession is mainly due to radiative trion recombination. The precession signal shortly before pump incidence, at negative delays, results from mode-locked hole spins.^{20,24} From the dependence of this mode-locking signal amplitude on the delay between pump pulses the hole spin coherence time can be assessed. The measured value depends sensitively on parameters such as pump power or sample temperature.

Due to slight variations in these parameters we typically find variations of the coherence time from a few hundred ns up to a μ s (see also below).

To induce spin echoes we extend the experimental setup by adding a further, pulsed Ti:Sapphire laser that is used for optical spin rotations.²⁵ This laser is synchronized to the pump-probe laser with an accuracy of 1 kHz. The rotation-pulse (RP) duration is also 1 ps. The RP repetition period, however, was taken either as emitted from the laser so that the pulses are separated by $T_{RP} = 13.2$ ns, or the pulse period was reduced to 6.6 ns by splitting the laser output into two pulse trains, sending one train along a mechanical delay line of 2 m length and reuniting it with the other pulse train.

The goal of this pulse train is to invert the momentary orientations of the precessing hole spins at a certain moment after initial orientation. In our case this moment is chosen such that the macroscopic hole spin coherence is already dephased. To reverse the dephasing, again excitation of the trion transition by circularly polarized pulses is applied. To avoid generation of new spin coherence, but manipulate existing spin coherence only, the area of these RPs is adjusted to $\Theta = 2\pi$. Under these conditions the system is excited during the pulse action to a positively charged trion and returned back to the resident hole. After this complete Rabi-flop the hole has acquired a geometrical phase, that corresponds to a rotation about the optical axis by an angle of π for resonant excitation.²⁶

As the RP is applied when the spin ensemble is dephased, the hole spin orientations are arbitrarily distributed in the plane normal to the magnetic field. In this case the RP action is identical to a reflection of the spins at the plane spanned by the optical axis and the magnetic field. Then, the precession after inversion brings

the dephased spins into phase again at time $2\Delta t$ that is twice the separation between rotation and pump pulses Δt . At this moment, the hole spins are again fully aligned (in case of perfect RP action on the ensemble, similar to pump pulse application), so that a further RP coming in later can induce a spin echo again. Since $T_{RP} \ll T_R$ in this experiment, the spins are rotated multiple times between two pump pulses. This leads to a sequence of dephasing and rephasing, resulting in multiple echoes between two pump pulses.

The RP and echo sequences between two subsequent pumps are shown schematically in Fig. 1(b). The upper part gives the case in which the RP separation is 13.2 ns, as indicated by the shaded green pulses equidistantly spaced in time relative to the black colored pump pulses. For simplicity, the separation between the pump pulses was assumed to be $T_R = 79.2$ ns, much smaller than the minimum separation in experiment, for clarity of discussion. The upper blue curve shows the expected signal echo sequence as result of the RP application. After pump incidence at zero delay the first RP hits at 12.1 ns, leading to echo formation at 24.2 ns delay. The next RP comes in 1.1 ns later at 25.3 ns delay and manipulates on the spin coherence from the previous echo, so that it induces the next echo another 1.1 ns later at 26.4 ns. The situation at this echo time is basically identical to that at the moment of pump pulse application so that the subsequent RPs repeatedly induce identical echo sequences.

Note that due to the particular periodicity of rotation and pump pulse application, a peculiar symmetry of the echo appearance in time arises. With the RP incidence at Δt , the echo appearance occurs at $2\Delta t$, independent of the sign of Δt . Furthermore, the echoes occur symmetrically to each RP. This can both be seen in the lower part of Fig. 1(b) showing the case when the RP period is bisected as indicated by the additional unshaded green pulses. Consequently the number of echoes is doubled with the additional echoes appearing just in between the ones in the previous case.

In the experiments the length of our mechanical delay lines for adjusting the different pulse trains relative to each other allows us to cover a delay range of about 6 ns, which therefore has to be selected carefully. We have decided for the range from about -4 ns to 2 ns. In both protocols with 13.2 ns and 6.6 ns pulse separation, respectively, a RP comes in at $\Delta t = -1.1$ ns, so that an echo (induced by a previous RP) should appear at $2\Delta t = -2.2$ ns. The corresponding experiments are shown by the two lower curves in Fig. 1(a). The green arrows indicate the moment in which the RP hits the sample in the two cases with different T_{RP} . In agreement with the expectations, hole spin echoes are observed at -2.2 ns in the ellipticity measurements. The echoes have comparable amplitude and show a similar fast dephasing as the hole spin coherent signal after a pump pulse. The echo character is confirmed by varying the moment of RP arrival Δt , as done in Fig. 2. When changing Δt from

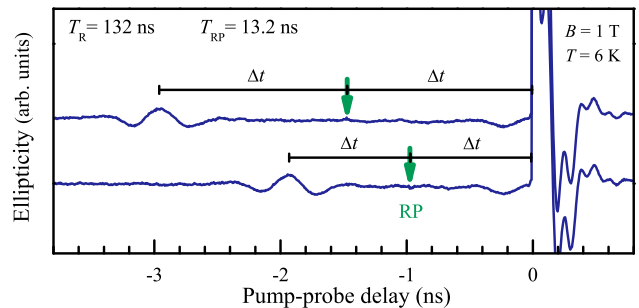


FIG. 2. (Color online) Spin echo emergence at time $2\Delta t$ for different arrival times Δt of the RP, -1.5 ns and -1 ns, as indicated by the green arrows before pump pulse arrival at zero delay. $B = 1$ T at $T = 6$ K.

-1 ns to -1.5 ns, the echo at $2\Delta t$ shifts correspondingly from -2 ns to -3 ns.

The RP train sequences are similar to the CPMG protocol originally implemented in NMR studies.^{12,23} This protocol has recently been used to keep a quantum bit embedded into surrounding baths alive. Simply speaking, its impact can be understood such that the RPs invert the spins within time periods that are short compared to the effective coupling time to the baths so that the spins become decoupled from them. The periodic RP sequences used in our experiments allow us to assess the potential of optical dynamic decoupling protocols for extending spin coherence.

To measure the hole spin coherence time, the period between two pump pulses T_R is increased using the pulse picker and the amplitude of the last echo before pump incidence at -2.2 ns delay is measured as a function of T_R . For reference, also the hole-spin mode-locking amplitude right before zero delay as a function of the separation between pump pulses without RP application is shown in Fig. 3(a). These data were recorded under the same experimental conditions (pump power etc), optimized for the longest possible coherence time achievable, as in the echo studies. The data are normalized to unity for zero delay. Within the scanned range of pump separations, the mode-locking amplitude decays due to the loss of coherence. To assess the underlying coherence time quantitatively we fit the data for the mode-locking amplitude $A(t = T_R)$ by an exponential decay. In detail, we use the fit form that was elaborated in Ref. [27] for mode-locked spins:

$$A(t) = A_0 \exp \left[- \left(2 + \frac{1}{2\sqrt{3} + 3} \right) \frac{t}{T_2} \right], \quad (1)$$

with the hole-spin coherence time T_2 . Due to the normalization of the data, $A_0 = 1$. From this fit we obtain a hole spin coherence time of $T_2 = 560$ ns, in reasonable accord with previous reports.^{20,22} This value serves as reference value for all subsequent experiments involving spin echo signals. For that purpose we accurately determine in every experiment the hole-spin mode-locking

amplitude and normalize it to this reference value to get rid of possible variations in experimental parameters.

The amplitudes of the last echoes before pump incidence induced by the two sequences of RP application with $T_{RP} = 13.2$ ns and $T_{RP} = 6.6$ ns are plotted as a functions of the time after the last pump incidence in the panels (b) and (c) of Fig. 3, respectively. This time is given by $t = T_R + 2\Delta t$ with $\Delta t = -1.1$ ns. Again, we have normalized the two data sets such that right after pump action and before the RPs can have an effect, the amplitudes are identical. This normalization has no impact on the exponential decay times. Also here one can see the decay of the echo amplitude with increasing pump separation T_R , but clearly the drop occurs much faster when the RP sequence with $T_{RP} = 13.2$ ns is applied compared to the 6.6 ns case. This indicates that indeed the coherence of the hole spins is kept alive more efficiently for short RP separation. We use the same fit form as for the mode-locked signal to assess the coherence time in these experiments. For the case with 13.2 ns RP separation we obtain a coherence time of $T_2 = 680$ ns from the fit of the experimental data which is only slightly longer than for the case without rotation pulses. When the separation between RPs is bisected, the coherence time increases by about a factor of two up to 1190 ns, more than twice the time without rotation pulses.

This result clearly supports decoupling from surrounding baths by periodic laser pulses. The limitation of the hole spin coherence at $T = 2$ K with a quick drop for temperatures higher than liquid helium has been tentatively assigned to the hyperfine interaction with the nuclei.²² On the other hand, this interaction has been shown to be about an order of magnitude weaker than for electrons,^{28–32} even though the observed spin coherence times are very much comparable for electron and hole spins.^{20,21} This underlines that further work needs to be done in this field to understand the coupling of spins to baths on a microscopic level. Dynamic decoupling techniques might be helpful in this respect: While the mode-locking data [Fig. 3(a)] can be quite well described by an exponential decay, the data recorded with RP application indicate that the echo amplitude decrease does no longer follow a pure exponential function but appears more complicated with an initial somewhat faster drop followed by a much slower decrease. By fitting the data points for times exceeding 200 ns, decay times exceeding 10 μ s are obtained. Further studies are required to understand this behavior in more detail.

In summary, we have demonstrated the potential of all-optical protocols for decoupling the dynamics of QD-confined carrier spins from the surrounding baths. While much shorter pulse durations can be obtained in that way, the main obstacle at the moment is the less accurate and flexible possibility to tailor optical pulses compared to the microwave range. However, the huge progress in pulsed laser technology might open novel perspectives here. This progress may be used to implement more complex dynamic decoupling protocols that are optimized for

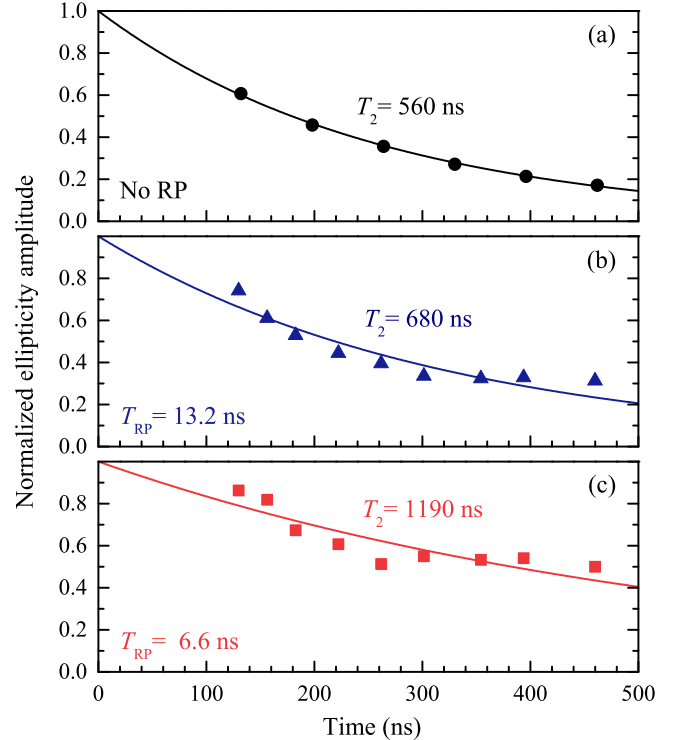


FIG. 3. (Color online) (a) Normalized ellipticity amplitude of mode-locked hole-spins right before a pump pulse in dependence of the time after previous pump incidence $t = T_R$ without RP application. (b) Normalized echo amplitudes in dependence of the time after previous pump incidence $t = T_R + 2\Delta t$ with $\Delta t = -1.1$ ns for a RP separation of $T_{RP} = 13.2$ ns. (c) Normalized echo amplitudes for $T_{RP} = 6.6$ ns.

keeping the coherence of a quantum bit alive.

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